

AMENDMENTS TO THE CLAIMS

1. (Currently amended) A process for ~~the synthesis of a~~ preparing an α -substituted acrolein ~~of~~ having the formula $\text{CH}_2=\text{CR}-\text{CHO}$ ~~where, wherein~~ R is an alkyl group or an aryl group, ~~the process~~ comprising:

(i) ~~subjecting a mixture of an olefin and syngas or a mixture of olefin and~~ an olefin, carbon monoxide and hydrogen to hydroformylation in an organic phase ~~and~~ in the presence of a hydroformylation catalyst or catalyst precursor ~~comprising that is~~ a rhodium complex catalyst or catalyst precursor to obtain the corresponding aldehyde; and

(ii) ~~subjecting the aldehyde obtained in step (i) above to aldol condensation with~~ formaldehyde in an aqueous phase in the presence of an aldolisation catalyst ~~comprising that is a~~ secondary amine or a secondary amine/organic acid catalyst ~~in aqueous phase~~,

wherein the reactions of steps (i) and (ii) ~~being~~ are carried out simultaneously in a biphasic aqueous-organic system, ~~ensuring the segregation of and the two catalysts~~ hydroformylation catalyst and the aldolisation catalyst are segregated in the into two distinct phases, ~~to obtain an~~ α -substituted acrolein.

2. (Currently amended) A ~~The process as claimed in~~ of claim 1, wherein the rhodium complex catalyst ~~comprises an organometallic complex of rhodium of~~ has the formula $\text{Rh}(\text{A})_p(\text{B})_q(\text{C})_r(\text{D})_s$, $\text{Rh}(\text{A})_p(\text{B})_q(\text{C})_r(\text{D})_s$, wherein A, B, C and D are ~~ligands~~ each independently selected from the group consisting of H, a trialkyl ~~phosphines~~ phosphine, a triaryl ~~phosphines~~ phosphine, carbonyl, a trialkyl arsine, a triaryl ~~arsines~~ arsine, an alkyl-aryl ~~phosphines~~ alkylaryl phosphine, a trialkyl ~~amines~~ amine, a triaryl ~~amines~~ amine, an alkyl-aryl ~~amines~~ alkylaryl amine, a ~~bisphosphines~~ bisphosphine and a

~~diimines diimine, and the sum of p+q+r+s~~ p and q and r and s is equal to 4, 5, or 6, ~~such that and the resultant complex catalyst~~ is soluble in the organic phase.

3. (Currently amended) A ~~The process as claimed in~~ of claim 1 wherein the organic phase ~~comprises an organic media~~ is selected from the group consisting of ~~an aromatic hydrocarbons hydrocarbon, an aliphatic hydrocarbons hydrocarbon, a higher alcohols alcohol, and any mixtures thereof.~~

4. (Currently amended) A ~~The process as claimed in~~ of claim 1 wherein the rhodium complex catalyst is selected from the group consisting of $\text{HRh}(\text{CO})(\text{PPh}_3)_3$, $\text{Rh}(\text{CO})(\text{Cl})(\text{PPh}_3)_2$, $\text{Rh}(\text{CO})_2(\text{acetylacetonate})$, $\text{HRh}(\text{CO})_2(\text{acetylacetonate})$, $[\text{Rh}(\text{Cyclooctadiene})\text{Cl}]_2$, ~~$\text{HRh}(\text{CO})(\text{Pn-butyl})_3$~~ , $\text{HRh}(\text{CO})(\text{P}(\text{n-butyl})_3)_3$, $\text{HRh}(\text{CO})(\text{PPh}_3)(\text{diphenylphosphinoethane})$, $\text{HRh}(\text{CO})(\text{PPh}_3)(\text{diphenylphosphineopropane})$, $\text{HRh}(\text{CO})(\text{PPh}_3)(\text{diphenylphosphinopropane})$, and $\text{HRh}(\text{CO})(\text{PPh}_3)(\text{diphenylphosphinobutane})$, $\text{HRh}(\text{CO})(\text{PPh}_3)(\text{diphenylphosphinobutane})$.

5. (Currently amended) A ~~The process as claimed in~~ of claim 1 wherein ~~the step of hydroformylation reaction (i) is carried out using in the presence of~~ a rhodium catalyst precursor with having an additional coordinated ligand and ~~in the presence of~~ an additional free ligand that is not coordinated to the rhodium metal, which wherein the additional free ligand is identical to or different from the additional coordinated ligand ~~already coordinated to the rhodium metal precursor.~~

6. (Currently amended) A ~~The process as claimed in~~ of claim 5 wherein the additional coordinated ligand is and the additional free ligand are water insoluble and is are each independently selected from the group consisting of an aryl secondary phosphine, an alkyl secondary phosphine, and an alkylaryl secondary phosphines phosphine, an aryl tertiary phosphine, an alkyl tertiary phosphine, and an alkylaryl tertiary phosphines phosphine, an aryl secondary

phosphate, an alkyl secondary phosphate, and an alkylaryl secondary phosphates phosphate, an aryl tertiary phosphate, an alkyl tertiary phosphate, and an alkylaryl tertiary phosphates phosphate, an aryl secondary arsine, an alkyl secondary arsine, and an alkylaryl secondary arsines arsine, an aryl tertiary arsine, an alkyl tertiary arsine, and an alkylaryl tertiary arsines arsine, an aryl tertiary amine, an alkyl tertiary amine, and an alkylaryl tertiary amines amine, a pentanediones pentanedione, a substituted pentanediones pentanedione, an acetylacetonates acetylacetonate, a Schiff bases base, and an aryl bisphosphine, an alkyl bisphosphine, and an alkylaryl bisphosphines bisphosphine.

7. (Currently amended) A The process as claimed in of claim [[5]] 6 wherein the additional coordinated ligand and the additional free ligand is are each independently selected from the group consisting of triphenylphosphine, tri n-butylphosphine, acetylacetonate, tributylphosphite, tributylphosphite, triphenylphosphite, triphenylamine, tributylamine and triphenylarsine.

8. (Currently amended) A The process ~~as claimed in~~ of claim 5 wherein the reaction (i)⁴ is carried out in presence of excess free ligand and a mole ratio of rhodium metal to free ligand is in the range of 0.1 to 10.

9. (Currently amended) A The process as ~~claimed in~~ of claim 1 wherein the aldolisation catalyst is soluble in the aqueous ~~medium~~ phase and ~~comprises~~ is a secondary aryl or alkyl amine selected from the group consisting of diethyl amine, dimethyl amine, ~~methyle~~ methyle ethyl amine, ~~dibutyl~~ dibutyl amine, dibenzyl diphenyl amine, piperidine, morpholine, piperazine and ~~pyrrolidine~~, pyrrolidine.

10. (Currently amended) A ~~The~~ process ~~as claimed in~~ of claim 1 wherein the aldolisation reaction catalyst is a secondary amine/organic acid catalyst and ~~the is carried out in the presence of~~ an organic acid is selected from the group consisting of acetic acid, propionic acid and butyric acid.

11. (Currently amended) A ~~The process as claimed in~~ of claim 10 wherein the mole ratio of the secondary amine aldolisation catalyst to the organic acid is in the range of 0.01 to 10.
12. (Currently amended) A ~~The process as claimed in~~ of claim 1 wherein the mole ratio of the hydroformylation catalyst to the aldolisation catalyst is in the range of 0.01 to ~~40000~~ 10,000.
13. (Currently amended) A ~~The process as claimed in~~ of claim ~~[[1]]~~ 12 wherein the mole ratio of the hydroformylation catalyst to the aldolisation catalyst is in the range of 0.1 to ~~4000~~ 1,000.
14. (Currently amended) A ~~The process as claimed in~~ of claim 1 wherein the organic phase ~~of the present invention~~ is immiscible with water and is selected from the group consisting of toluene, xylene, cyclohexane, heptane, decanol and ~~any~~ a mixture thereof.
15. (Currently amended) A ~~The process as claimed in~~ of claim 1 wherein the formaldehyde is ~~used~~ in the form of an aqueous solution or ~~in the form of~~ paraformaldehyde.
16. (Currently amended) A ~~The process as claimed in~~ of claim 1 wherein the mole ratio of olefin to formaldehyde is in the range of 0.1 to 100.
17. (Currently amended) A ~~The process as claimed in~~ of claim ~~[[1]]~~ 16 wherein the mole ratio of olefin to formaldehyde is in the range of 0.5 to 10.
18. (Currently amended) A ~~The process as claimed in~~ of claim 1 wherein the olefin is ~~selected from a substituted and or unsubstituted olefins with a~~ olefin having from 2 to 10 carbon number from 2 to 10 atoms.
19. (Currently amended) A ~~The process as claimed in~~ of claim 1 wherein ~~the reaction~~ reactions (i) and (ii) are carried out at a temperature ~~varies between 20 °C to and~~ between 20 °C and 200 °C.
20. (Currently amended) A ~~The process as claimed in~~ of claim 1 wherein the ~~pressure~~ pressures of carbon monoxide and hydrogen ~~is~~ are each independently in the range of 10 to 1500 psi ~~each~~.

21. (Currently amended) A ~~The process as claimed in~~ of claim ~~[[1]]~~ 20 wherein the ~~pressure pressures~~ of carbon monoxide and hydrogen ~~is~~ are each independently in the range of 50 to 450 psi each.
22. (Currently amended) A ~~The process as claimed in~~ of claim 1 wherein the ~~product α~~ substituted acrolein product is preferentially soluble in the organic ~~media~~ phase.
23. (Currently amended) A ~~The process as claimed in~~ of claim 1 wherein ~~the reaction is~~ reactions (i) and (ii) are conducted either as a batch or as a continuous reaction wherein the ~~with~~ continuous addition of olefin, and carbon monoxide, and hydrogen are added continuously at a rate that is dependent on their rate of consumption ~~dependant on consumption rate thereof~~.
24. (Currently amended) A ~~The process as claimed in~~ of claim 1 wherein the olefin ~~used~~ is ethylene and the α -substituted acrolein ~~product obtained~~ is methacrolein.